

Evaluation of In Situ Grouting for Operable Unit 7-13/14

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ABSTRACT

The U.S. Department of Energy is conducting the Waste Area Group 7 Operable Unit 13/14 Comprehensive Remedial Investigation/Feasibility Study at the Idaho National Engineering and Environmental Laboratory to satisfy requirements of the Federal Facilities Agreement and Consent Order with the State of Idaho and the U.S. Environmental Protection Agency. The Comprehensive Environmental Response, Compensation and Liability Act governs these activities, which involve assessing contaminants of concern, risk factors, and potential technologies for remediation.

This report describes the technology of in situ grouting as applied to treatment of buried waste at the Subsurface Disposal Area within Waste Area Group 7. This document presents currently available technology performance information and serves as a reference document in the pending feasibility study.

Discussions in this report summarize applying in situ grouting to radioactively contaminated waste and soil sites across the United States and reports technology performance data where available. One analysis discusses different in situ grouting techniques to determine suitability for stabilizing or treating waste buried at the Idaho National Engineering and Environmental Laboratory. In addition, this document presents an analysis of available data to determine effectiveness of the technology at inhibiting release of contaminants from the Subsurface Disposal Area as well as durability of the resulting waste form over time.

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ACRONYMS

ANS	American Nuclear Society
ANSI	American National Standards Institute
COC	contaminant of concern
DOE	U.S. Department of Energy
DOE-ID	U.S. Department of Energy Idaho Operations Office
DUST	Disposal Unit Source Term (computer model)
INEEL	Idaho National Engineering and Environmental Laboratory
ISG	in situ grouting
OU	operable unit
RFP	Rocky Flats Plant
RWMC	Radioactive Waste Management Complex
SDA	Subsurface Disposal Area
STD	standard
TRU	transuranic
VOC	volatile organic compound
WAG	waste area group

Evaluation of In Situ Grouting for Operable Unit 7-13/14

1. INTRODUCTION

The U.S. Department of Energy (DOE) is conducting the Waste Area Group (WAG) 7 Operable Unit (OU) 13/14 Comprehensive Remedial Investigation/Feasibility Study within the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory (INEEL) to satisfy requirements of the Federal Facilities Agreement and Consent Order (DOE-ID 1991) with the State of Idaho and the U.S. Environmental Protection Agency. The Comprehensive Environmental Response, Compensation and Liability Act (42 USC § 9601 et seq.) governs these activities, which involve assessing contaminants of concern (COCs), risk factors, and potential technologies for remediation.

1.1 Purpose and Scope

The in situ grouting (ISG) technology has been widely investigated as a potential solution to the challenges of buried waste facing DOE. Tested in simulated environments and applied successfully at a number of actual remediation sites, ISG provides a mechanism to isolate potentially harmful chemicals from the environment, reduce contaminant mobility, and provide overall protection of human health and the environment for extremely long periods of time. In addition, the technology decreases risk associated with remediation, thus protecting workers by avoiding direct contact with hazardous waste.

This report maintains a narrow focus on the application of ISG as a remediation technology for mixed radioactive waste landfills. The document evaluates effectiveness and implementability of the technology and summarizes previous applications of ISG. This document is intended to provide technology performance data for the OU 7-13/14 feasibility study at the Subsurface Disposal Area (SDA) within the RWMC (i.e., WAG 7).

1.2 Technology Description

In situ grouting was developed in the construction industry and recently adapted for environmental use. The process entails injecting a slurry-like mixture of cements, chemical polymers, or petroleum-based waxes into contaminated soil or a waste landfill. Grouts are specially formulated to encapsulate contaminants and isolate them from the surrounding environment. In the environmental industry, the process is described as nondisplacement jet grouting where soil and waste debris are mixed subsurface, forming a large grout monolith (DOE 1999a; Loomis, Zdinak, and Bishop 1997). In situ grouting also has been investigated as a way to construct barriers around waste pits or contaminated areas. Grouts have been used successfully to form sidewalls and under-barriers around contaminated areas. However, the size of the SDA and the shallow basalt bedrock poses problems for constructing under-barriers. Therefore, this report focuses on applying ISG to create monoliths in the waste zone.

Grout typically is pumped into the waste zone under pressure using some form of injection lance. Injection lances are inserted into the waste zone using hydraulic hammers or modified drill rigs in a tightly spaced injection pattern. Grouting is accomplished without displacing contaminants or debris or ground heaving. The overall site volume remains constant, but density of the site is increased substantially.

The injection method produces interlocking columns of grout extending from the underburden soil up through the waste, terminating subsurface in the overburden. The interlocking columns cure into a solid monolith with no discernable edges between columns. Containers of waste are filled from the inside with grout. When injected under high pressure, the cutting action of the jets fractures soil, plastics, wood, and other low-strength objects. Cutting action of the jets dislodges particles and small pieces of waste material and mixes them with grout and soil. Small amounts of liquid in the waste are impacted by the grout stream and dispersed randomly over short distances in the grout. Large objects remain in place as the grout flows under pressure into voids around objects. All readily accessible voids are filled (Loomis, Zdinak, and Bishop 1997).

When properly designed and applied, ISG produces a durable waste form resistant to weathering and degradation over long periods of time. In situ grouting reduces mobility of contaminants by the following mechanisms:

- **Reduced permeability:** Injecting grout under high pressure into the disposal area fills void space around debris objects and in the soil matrix. Properly spaced injection points will rupture waste containers and fill void space inside waste drums and boxes. The resultant grout and waste monolith has a low porosity and hydraulic conductivity.
- **Stabilization:** Substantially reduced void space in the waste and soil matrix prevents future compaction and subsidence of the pits. An incompressible foundation is a critical component for durable cover systems.
- **Encapsulation:** Energetic mixing of grout with waste on the inside of containers and with soil on the outside of containers encases contaminants in a leach-resistant matrix and minimizes the potential for contaminants to be mobilized by infiltrating water.
- **Chemical buffering:** An appropriately selected grout will chemically alter infiltrating water to reduce the solubility potential of contaminants. In addition, many grouts exhibit an affinity for specific contaminants and chemically can bind contaminants to reduce leachability.

In situ grouting waste forms may be expected to endure for thousands of years without significant physical or chemical alteration. Because the grout monolith is constructed 4 to 5 ft below ground surface, it is protected from many degrading mechanical forces (e.g., freeze and thaw cycles). Selecting grouts that are in chemical equilibrium with site-specific geochemistry minimizes degrading chemical forces. Though some cracking is expected as grout cures, release of contaminants is limited by chemical properties of the grout and infiltrating moisture. The primary degrading mechanism is natural dissolution of the grout matrix by infiltrating soil moisture. Specifically, formulating grouts that have natural analogs (long-lived naturally occurring materials) would allow waste forms to be constructed that would dissolve only over geologic time.

1.3 State of Development

In situ grouting activities have been performed at all major DOE sites using a variety of technical approaches. As early as 1985, DOE was evaluating grouting as a viable remedial technology for buried waste and contaminated soil sites (EG&G 1985). In situ grouting has been selected as a remedial action by DOE and its regulators and has been implemented successfully at several waste sites including:

- Oak Ridge National Laboratory in 1996
- Idaho National Engineering and Environmental Laboratory in 1997

- Brookhaven National Laboratory in 1999
- Savannah River Site in 2000.

In situ grouting has not been applied on a large scale to sites such as the SDA, but extensive research at the INEEL using simulated buried waste pits has been conducted to evaluate the efficacy of ISG. As a result, the implementability of ISG equipment and processes is well understood for buried waste pits. Interactions and limitations presented by specific waste forms found at the SDA have been investigated in numerous bench scale and field scale tests simulating buried waste pits. The permeability and porosity of interstitial soils have been evaluated and effective grouting parameters (i.e., injection pressures, grout viscosities, and volumes) have been developed by trial and error.

The effectiveness of ISG is also well understood. Conceptually, it is clear that grouting would substantially fill void space and encapsulate contaminants, thereby reducing leaching from the waste. Though parameters such as hydraulic conductivity, chemical buffering, and monolith cracking have proven difficult to measure in field scale applications, a limited body of data does exist. In addition, the DOE has conducted a substantial amount of grout waste-form testing. Available laboratory and field scale data, though not complete, indicate that ISG would be an effective alternative to treat SDA waste.

2. BACKGROUND

The U.S. Atomic Energy Commission (now DOE) established the INEEL (originally called the National Reactor Testing Station) in 1949 as a site for building and testing a variety of nuclear facilities. The SDA comprises all property from the center of the RWMC westward and is surrounded by a soil berm and drainage channel. The site was initially established in July 1952 as the National Reactor Testing Station Burial Ground on 13 acres (5 ha). The facility was expanded incrementally over the years and now covers 97 acres (39 ha). Waste disposed of in the SDA included low-level radioactive, mixed, and transuranic (TRU) waste from on- and off-Site generators that was originally dumped into trenches and consisted of debris that included paper, laboratory ware, filters, metal, pipe fittings, and other items contaminated by mixed-fission products. The waste was typically packaged in cardboard boxes that were taped shut and collected in dumpsters. The dumpsters were then emptied into the trenches (Loomis, Jessmore, and Weidner 2001). Land disposal of mixed and TRU waste was discontinued in 1970.

2.1 Site-Specific Conditions

Geologic characteristics significantly influence both the selection of grout types and the method of injection. The SDA is located in the semiarid desert of southeastern Idaho where average annual precipitation is 22 cm (8.7 in.) and depth to the underlying Snake River Plain Aquifer averages 177 m (580 ft). Basalt lava flows and other volcanic features (e.g., cinder cones and pressure ridges) govern topography. During quiescent periods between volcanic events, sediments were deposited on the surface of the basalt. Irregular topography of the basalt flows caused sedimentary materials to accumulate in isolated depressions, and the deposits display a wide range of grain-size distributions correlating to the depositional mechanism.

Soil found near the SDA is shallow, consisting of fine-grained eolian soil deposits with some fluvial gravels and gravelly sands (EG&G 1988). Soil at the SDA is composed of clay, silt, and sand. Clay minerals predominate (50 wt%), with quartz (37.5 wt%), calcite (10 wt%), and iron oxyhydroxide and other minerals (2.5 wt%) comprising the remainder (Lee, Martins, and Weidner 1991).

Hydrogen potential in local soil is approximately 8 (± 0.5), buffered by calcite-water-CO₂ interactions. The oxidation-reduction potential (Eh) is an oxidizing reaction and equivalent to air. The soil moisture is saturated with calcite, and super-saturated with dolomite (Wood and Norell 1996). Caliche (CaCO₃) formation, an impermeable, concrete-like soil naturally cemented by calcite, is commonly observed in the INEEL soil (DOE 1999a). In the SDA waste area, however, hydrogen potential (pH) of soil water is found to be only slightly alkaline (ranging from 7.2 to 8.2), possibly caused by high concentrations of CO₂ in soil gas resulting from organic decomposition of waste materials (Hull and Pace 2000).

2.2 Waste Disposal

Types of waste being treated affect the implementability of ISG. This section presents a brief description of waste historically disposed of at the SDA, including types of contaminants and media affiliated with each waste stream. A detailed description of the waste is available in comprehensive inventories developed for the SDA (LMITCO 1995a, 1995b).

2.2.1 Disposal Operations

As noted previously, the SDA contains waste predominantly generated by DOE facilities. This waste originated from the Rocky Flats Plant (RFP),^a INEEL, and a variety of U.S. Department of Defense installations, as well as several commercial and other government generators approved by DOE.

Waste is buried in trenches, pits, and soil vaults that were excavated down to underlying bedrock. Depth to bedrock (i.e., thickness of the surface soil) ranges from 0.6 to 7 m (2 to 23 ft) within the SDA. Trenches were long excavations with an approximate width of 2 m (7 ft). Because trench widths did not accommodate large items, large pits were excavated for disposing of bulky waste and large waste shipments. Rows of soil vaults, augured holes 15 and 57 in. in diameter, were used for disposing of low-level waste with high radiation levels. Figure 1 illustrates waste areas distributed across the SDA.

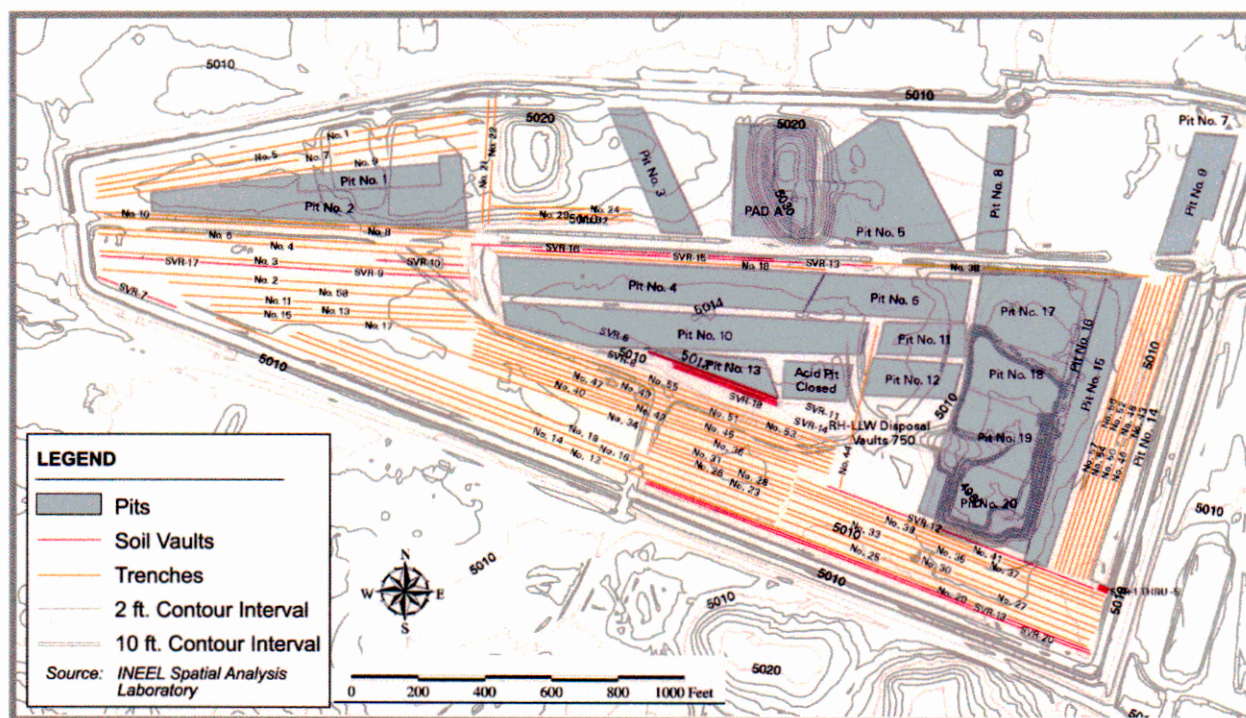


Figure 1. Waste areas distributed across the Subsurface Disposal Area within the Radioactive Waste Management Complex.

Waste could not be buried in some areas of the RWMC because surficial soil was too shallow. As a result, layout of the disposal areas in the landfill is highly irregular. In one area of the SDA (Pad A), drums and boxes of waste were placed on a surficial asphalt pad and bermed over with soil.

a. The Rocky Flats Plant is located 26 km (16 mi) northwest of Denver. In the mid-1990s, it was renamed the Rocky Flats Environmental Technology site. In the late 1990s, it was renamed the Rocky Flats Plant Closure Project.

A total of 20 pits, 58 trenches, 21 soil vault rows, and one aboveground pad were constructed in the SDA. Pits 1 through 10 and Trenches 1 through 51 accepted waste disposal from 1952 to 1969. The majority of the waste in Pits 1, 2, 4, 5, 6, 9, and 10 was generated by the RFP weapons production operations. Pad A was not used until 1972, and soil vault disposal began in 1977.

2.2.2 Waste Types and Containers

Waste in the SDA consists primarily of RFP disposals, irradiated metals, and contaminated debris. Waste contaminants include radioactive and nonradioactive hazardous materials such as volatile organic compounds (VOCs), mercury, beryllium, asbestos, zirconium fines, solidified acids and bases, solvents and degreasing agents, and sodium and potassium nitrates. Waste was disposed of in loose form or packaged in a variety of containers such as steel drums, and wooden and cardboard boxes. The following types of waste materials are present in pits and trenches throughout the SDA:

- Contaminated soil throughout all pits and trenches including soil underburden, overburden, and interstitial soil.
- Buried debris waste packages including boxes, drums, concrete blocks, and metal waste
- Buried drums of mixed TRU and low-level sludge waste including inorganic (water-based) sludge, organic (oil-based) sludge, and nitrate salts
- Liquid waste placed in the RWMC Acid Pit.

As discussed in subsequent sections, drums containing Series 743 and 745 sludge from the RFP are particularly important. Series 743 sludge consisted of approximately 30 gal of organic liquid stabilized with 100 lb of calcium silicate and 10 to 20 lb of absorbent. Oil in the sludge is primarily Texaco Regal, but also includes hydraulic, gearbox, and other mineral oils of similar density. Series 745 sludge originated from evaporating process liquids in solar ponds at the RFP. Residual sludge was dried and placed in 55-gal drums. Sludge consists of approximately 60% sodium nitrate, 30% potassium nitrate, and 10% miscellaneous materials.

3. EFFECTIVENESS

Remedial action performance history does not exist for a site as large and complex as the SDA. As with other alternatives, no substantial body of real-world data can be examined to understand the effectiveness of ISG to the SDA. Effectiveness must be evaluated on engineering estimates of performance under SDA conditions. For in situ treatment alternatives, effectiveness is measured by the waste form's ability to control the rate that contaminants are released to the environment. An effective waste form will release contaminants at a slow enough rate that any potential for future groundwater contamination and associated health risks are extremely low and acceptable under regulatory guidelines. For ISG, the processes that primarily control the contaminant release rate are permeability (hydraulic conductivity), diffusion, solubility, and chemical fixation. By using available literature, data, and a computer model (e.g., WAG 7 Disposal Unit Source Term [DUST] release model) (Becker et al. 1998), a conservative estimate of the ISG release rate may be made.

3.1 Hydraulic Conductivity

Water spreading contamination from buried waste poses the primary potential human health and ecological risk. Because no shallow groundwater is found at the SDA, infiltrating precipitation provides the primary mechanism to mobilize contaminants downward through the vadose zone. Grouting has been used in a variety of applications to reduce hydraulic conductivity, thereby reducing the flow of water through the site. Applying grout to waste sites has been shown to reduce hydraulic conductivity, which at the SDA would minimize the amount of moisture in contact with waste. Average hydraulic conductivity demonstrated for grouted sites is approximately 10^{-7} cm/second, equivalent to permeability typically achieved with engineered liners at hazardous waste landfills (Fetter 1994).

Hydraulic conductivity of grout matrices has been measured on a laboratory scale using methods such as American Society for Testing and Materials D-5084 (Milan et al. 1997). Hydraulic conductivity measured on bench scale samples of grout is consistently low (10^{-7} to 10^{-11} cm/second) for a wide variety of grout and soil mixtures (Heiser and Dwyer 1997). Average permeability measured in the laboratory on mixtures of different grouts and INEEL soil, sodium nitrate, and canola oil (simulations of SDA waste) met instrument detection limits at 10^{-11} cm/second (Milan et al. 1997).

Field scale hydraulic conductivity data are limited. Several field scale tests, including those at Oak Ridge National Laboratory and INEEL, indicate that measuring permeability in buried waste cells is complex and difficult. Results from the INEEL field scale permeability tests were inconclusive because fully saturated conditions never were achieved before the field season ended. However, double packer testing in core holes indicated an average hydraulic conductivity less than 10^{-7} cm/second (Loomis, Zdinak, and Bishop 1997). Measurements conducted at Oak Ridge National Laboratory WAG 4 yielded an average hydraulic conductivity of 0.9×10^{-6} cm/second (ORNL 1993; 1997). At Brookhaven National Laboratory, hydraulic conductivity measurements for cores taken from the cement layer ranged from 1.1×10^{-6} cm/second to 1.6×10^{-8} cm/second and averaged 3.4×10^{-7} cm/second. At the Sandia National Laboratory, measurements on field scale tests identified conductivities ranging from 8.4×10^{-7} to 2.7×10^{-9} cm/second, depending on grout type (Dwyer 1994).

Available field scale data make it difficult to discern whether measurements of hydraulic conductivity were representative of actual permeability of the waste and grout matrix on a micro scale, or whether results were influenced by fracture flow or edge effects. In cases where the field scale hydraulic conductivity is a result of fracture flow, the majority of contaminants are still encapsulated in grout and do not interact with water. Though treatability studies are planned at the INEEL to differentiate these effects, definitive data do not currently exist. From a cursory evaluation of literature data and field scale

testing, it is apparent that hydraulic conductivity of the in situ grout matrix can be expected to be less than 1.0×10^{-7} cm/second for most types of cementitious grouts. Hydrocarbon-based grouts (e.g., waxes) are virtually impermeable to water; bench-scale conductivity tests on Waxfix fell below instrument detection limits at 2×10^{-11} cm/second (Milan et al. 1997). As a point of comparison, an ungrouted (baseline) simulated waste site showed permeability of roughly 1.0×10^{-5} cm/second (Loomis, Zdinak, and Bishop 1997).

3.2 Contaminant Solubility

Leaching of contaminants also depends on chemical processes affecting concentration of contaminants in pore water in the waste zone. Solubility is the total amount of a mineral that can remain in solution under a specified set of conditions in the presence of an excess of the mineral in solid phase (Kemper 1990). The following will control contaminant solubility:

- Concentration of other chemical species in the water (natural minerals and contaminants)
- Mineral phase
- Hydrogen retention of the solution
- Oxidation-reduction potential of the solution.

Solubility at the SDA can be thought of in terms of ongoing natural processes. Observations of caliche deposits in the soil and vadose zone around the SDA is evidence that near-surface water is saturated with naturally occurring minerals and is depositing calcium, silica, and other minerals as it travels through the vadose zone (Weidner et al. 2000).

Hull and Pace (2000) calculate solubility for each of WAG 7 COCs under varying geochemical conditions. Results indicate that pH, and to a lesser extent Eh, contribute to wide variability in solubility. In general, a moderately reducing environment with pH ranging from 7 to 8.5 would minimize the solubility of most SDA COCs. Current SDA water and soil conditions may be near optimum to minimize solubility of contaminants (Hull and Pace 2000). Realistic modeling of contaminant release must consider chemical properties of actual SDA water and the fact that solubility potential for COCs is very low under SDA geochemical conditions. Selecting grouts compatible with these conditions will help minimize release rates. Ongoing studies at the INEEL are working to measure actual geochemical conditions of candidate grouts, and develop release rate simulations that better account for contaminant solubility. Expected results were published in the *Final Results Report, In Situ Grouting Technology for Application in Buried Transuranic Waste Sites* (Loomis et al. 2002). However, for the feasibility study, release rates will be estimated conservatively using the solubility limit of each contaminant in deionized water.

3.3 Chemical Fixation

Diffusion data, typically obtained using the American Nuclear Society and American National Standards Institute (ANS/ANSI) procedure, *American National Standard Method for the Measurement of the Leachability of Solidified Low-Level Radioactive Wastes by a Short Term Test Procedure* (ANS/ANSI-16.1), assume that the concentration of contaminants and other competing minerals in the leachant is zero. This provides the most conservative, albeit unrealistic, scenario. Though hydraulic conductivity often is considered a controlling parameter in leach rates, chemical interactions between the contaminant, the grout, and the infiltrating water have been shown to play a greater role in many cases. In addition to providing buffering effects on intergranular water, a number of grouts have been shown to

chemically fix contaminants in a mineral phase through several mechanisms, including elemental substitution and conversion of the contaminant to an insoluble form (Singh et al. 1997). In recent leach tests conducted at the University of Akron, Ohio (Loomis et al. 2002), phosphate-based grout exhibited a hydraulic conductivity of 1.5×10^{-7} cm/second, an average of two orders of magnitude higher than other grouts tested. However, that grout also exhibited one of the lowest leach rates of all grouts tested (1×10^{-14} to 1×10^{-15} cm²/second for strontium). These results provide strong evidence of chemical fixation.

Past work with actual waste similarly has provided evidence of substantial chemical immobilization. In a number of cases at the INEEL and Oak Ridge National Laboratory, mixtures of grout and actual waste were tested using the U.S. Environmental Protection Agency toxicity characteristic leaching procedure. Results indicated that high concentrations of hazardous metals (including mercury and lead) could be immobilized chemically, even to the point that the leachant fell below Resource Compensation and Recovery Act Universal Treatment Standards (Lewis et al. 2000; Loomis et al. 1998b). With the toxicity characteristic leaching procedure tests, samples are ground and then leached using deionized water. Because the test method destroys physical integrity of the grout, results are a good measure of chemical fixation.

3.4 Contaminant Diffusion

The dominant mechanism of release is determined by the hydraulic conductivity of the grout and waste matrix. In matrices with high hydraulic conductivity, contaminants are transported by advective flow. Alternatively, in low conductivity conditions, the primary mechanism is diffusion where contaminants are dissolved by and diffuse through relatively static intergranular water to edges of the monolith where infiltrating water transports the contaminant away from the disposal area.

Because of expected low hydraulic conductivity, the dominant release mechanism for ISG is considered diffusion. Fick's Law generally describes diffusive flux of a chemical specie through porous media, written in finite difference form (Kemper 1990) in Equation (1).

$$\Delta Q / \Delta T = A D_p (\Delta C / \Delta x) \quad (1)$$

where

ΔQ = amount of the component that has diffused in the time ΔT .

D_p = diffusion coefficient in porous media.

A = cross sectional area.

Δx = thickness of the diffusion media.

$\Delta C / \Delta x$ is equivalent to the concentration gradient in Fick's law. By assuming C is negligible outside the block, ΔC is equal to the concentration in the waste form.

The surface area (i.e., A) and thickness of the diffusion media (i.e., Δx) are dependent on the treatment application design. An ISG monolith could be as small as a single soil vault or as large as an entire pit. In addition, presence of fractures in the monolith effectively reduces the size of the block

(i.e., the diffusion calculation would apply to subblocks between fractures). For the feasibility study evaluation, a variety of block sizes can be evaluated to bound the expected release rates in the absence of fracture frequency and aperture data. For example, each 2-ft-diameter column of grout could be considered an individual waste block.

The concentration term (i.e., C) is difficult to predict realistically. Highly complex chemical reactions among the contaminants, soil gas, soil moisture, and dissolved minerals have a significant bearing on the rate and amount of contaminant that go into solution. When geochemical properties of the specific SDA grout are measured, solubility limits can be calculated. Until that time, very conservative estimates may be made by assuming that total mass of the COCs is in solution and uniformly distributed across the volume of the ISG monolith, as described by Sullivan (1993).

The diffusion coefficient (i.e., D_p) for low-level radionuclides has been routinely measured in cementitious waste forms. Typically, diffusion coefficient data are derived from short-term (90-day) bulk leach methods (e.g., ANS/ANSI-16.1). The standard test requires a monolithic sample (cylinder) and demineralized water leachant. The leachant is extracted and replaced at specified time intervals with new water. Given the geometry of the specimen and the leachant composition over time, the diffusion coefficient can be computed.

Diffusion is largely dependent on grout type and, to a lesser extent, on chemical species of the contaminant (Weidner et al. 2000). Unfortunately, only limited data are available for actinides, and virtually all data are specific to concrete waste forms.

Tests such as ANS/ANSI-16.1 were designed to provide relative indexing between waste forms, not to predict long-term leaching behavior. Defensible techniques to extrapolate short-duration test data extending tens of thousands of years into the future do not exist. A satisfying set of analytical data has not been, and could not have been, developed to support the feasibility study evaluation. However, data derived from ANS/ANSI-16.1 can be considered representative of *initial* leaching conditions of the ISG waste form. Evaluation of long-term leaching behavior will need to be based on an analysis of the physical and chemical stability of grout in the SDA environment.

As mentioned previously, conservative estimates of release can be made using computer models to calculate movement of contaminants. Though other methods can analyze release (e.g., using the water infiltration rate combined with a retardation factor as presented by Hull and Pace [2000]), release of contaminants from grout waste forms traditionally has been evaluated using diffusion coefficients derived from ANS/ANSI-16.1 or similar tests. Weidner (2000) summarizes diffusion coefficient data applicable to SDA COCs. Several example diffusion coefficients for Portland cement waste forms are shown in Table 1.

Table 1. Example diffusion coefficients for cementitious waste forms.

Element	Diffusion Coefficient (cm ² /second)
Carbon	10 ⁻¹⁴
Uranium	10 ⁻¹³ to 10 ⁻¹⁵
Plutonium	10 ⁻¹⁰ to 10 ⁻¹⁷

Because of past test results of grouts tailored to specific contaminants (Singh et al. 1997; Loomis, Zdinak, and Bishop 1997), grouts formulated specifically for the SDA would be expected to exhibit even greater leach indexes.

Additional diffusion data are available for certain contaminants. For example, during tests of grouting Oak Ridge National Laboratory underground storage tank sludge, Sr-85 and Cs-137 exhibited excellent leach resistance with leachability indexes greater than 10, as measured by the ANS/ANSI-16.1 leach test (Spence and Kauschinger 1997).

3.5 Release Rates

In application, specific grout formulation are designed to minimize leaching by chemical reaction or reduced hydraulic conductivity, depending on specific COCs. However, a modeling approach that uses literature diffusivity values and ignores the added benefits of solubility limits and chemical fixation would provide an extremely conservative estimate of release. Appendix A, "Preliminary Release Rate Modeling Results," provides a simple estimate of a release rate based on the diffusion model described above. One contaminant, uranium, was used as an example to demonstrate the basic process. Results of the simulation, using a 1-m³ block and a diffusion coefficient of 1×10^{-13} , resulted in a release rate of approximately 0.01 mg/year, roughly two orders of magnitude lower than the (untreated) base case.

In the feasibility study, additional model runs will be performed to evaluate additional COCs. However, using the ANS/ANSI-16.1 (Spence and Kauschinger 1997) data in the simple diffusion model is like submerging the SDA grout monolith in a slowly moving stream of deionized water. Though the model is not realistic, results provide a conservative estimate that may be useful when comparing release rates for multiple alternatives.

4. DURABILITY

Durability of grout materials can be evaluated by analyzing (1) mechanical and geochemical forces of weathering and (2) natural and fabricated materials demonstrating durability over history. Such analyses make it clear that in the SDA environment, some grout types are expected to remain physically and chemically unaltered over very long periods.

4.1 Degrading Forces

Degradation of grout in the natural environment involves the same weathering processes that affect the stability of rock (Weidner et al. 2000). Durability of all rock-like materials is determined by the impact of weathering processes on the material and resistance of the material to such forces over time. Mechanical processes that can affect grouts include compression from volume increases caused by salt crystal growth, freeze and thaw cycles, temperature changes, and wet-dry episodes. Other mechanical stresses are caused by compaction from the force of gravity and tension created by shrinkage. Chemical reactions with contacting water, soil, and air also contribute to weathering. Chlorides, sulfates, and low-pH conditions have significant reactions with cementitious materials and can cause rapid degradation. Finally, some types of microbial activity can create conditions that rapidly degrade concrete.

Analysis of the subsurface SDA environment reveals that very few forces are available to degrade the grout waste form. For example, because ISG is applied at depth, soil temperatures are virtually constant and freeze and thaw cycles will not affect the grout. Table 2, adapted from Weidner et al. (2000) summarizes the impact of specific forces on ISG as applied to buried waste at the SDA.

Table 2. Summary of the impact of degrading forces on in situ grouting as applied to the Subsurface Disposal Area.

Degrading Force	Impact on the Subsurface Disposal Area	Rationale
Biodegradation	Negligible	Bacteria species that degrade cementitious and hydrocarbon-based materials cannot survive in the SDA environment because of the high soil pH; low temperature; and absence of free sulfur, thiosulfates, and inorganic ferrous iron.
Freeze and thaw	Negligible	Waste and grout monolith is less than 6 ft below land surface. Temperatures are constant at approximately 60°F.
Wet and dry	Negligible	Intergranular water tension is constant, slightly less than saturation.
Compression and shear	Negligible	Waste and grout monolith is supported on five sides and available void volume is filled.
Temperature changes and elevated temperature	Negligible	Waste and grout monolith is less than 6 ft below land surface. Temperatures are constant at approximately 60°F.
Sulfide and chloride attack	Negligible	Sulfides and chlorides are not present naturally in the SDA environment and are present only in select SDA waste streams.

Table 2. (continued).

Degrading Force	Impact on the Subsurface Disposal Area	Rationale
pH and Eh	Grout dependent	The pH conditions in SDA water and soil are slightly alkaline (pH ~8), favorable for most grout materials. The Eh is equivalent to oxygen in air, which is also favorable for most grout materials.
Shrinkage and cracking	Low	Cracking is not well understood and largely dependent on the particular grout. However, the effect of cracking on contaminant migration is thought to be minor because of the encapsulating nature of grout. Using soft grouts virtually eliminates the potential for fractures because any cracks formed during curing would close under ambient pressure. Further study is needed to better understand the issue of cracking.
Water dissolution	Low	Because the SDA is in an arid region with very low infiltration rates, dissolution of grout minerals will occur at a very slow rate. Soil water in the SDA is saturated with respect to calcium and magnesium, and nearly saturated with silica. The SDA is a region of mineral precipitation, not leaching. The SDA chemical environment will not support rapid dissolution of grout components.

pH = hydrogen potential
 Eh = oxygen and reduction potential
 SDA = Subsurface Disposal Area

Very few mechanisms are available in the SDA region to chemically or physically alter grout waste forms. The two possible mechanisms (i.e., potential cracking and water dissolution) would be expected to produce effects only at extremely slow rates, similar to those observed for the weathering of natural rock minerals.

4.2 Historic Examples

Numerous examples exist of both naturally occurring and fabricated materials that have endured for long periods. Table 3 provides examples of fabricated cementitious materials that have proven to be durable for many years.

Table 3. Examples of durable cementitious materials.^a

Example Site	Cementitious Matrix
Mortar from the Great Pyramid, Giza, Egypt, 2500 BC	Gypsum cement
Lime-soil mix from the Great Wall, China, 100 to 200 BC	Lime-soil mix
Mortar from Mole, Pozzuoli, Italy, 100 to 200 BC	Pozzolonic lime cement
Roman Aqueduct, Lyon, France, 200 to 300 AD	Lime cements (aluminous, ferruginous, and siliceous)

a. Weidner et al. (2000).

Materials listed in Table 3 have survived because of the absence of, or negligible impact from, degrading forces such as those described above. These materials have remained essentially unaltered chemically or physically since their original formulation (Jiang and Roy 1993). Weidner (2000) suggested that the selection of appropriate grouts for the SDA should be based largely on their chemical compatibility with the surrounding environment, stating, “A grout at chemical equilibrium with the environment will not chemically degrade whatsoever, provided the climate and other factors of the environment do not significantly change over time.”

4.3 Expected Durability

Many forces that cause rapid degradation in familiar cementitious materials do not play a significant role in the SDA ISG application. A number of factors are grout dependent; therefore, deteriorating effects will occur only when selected grout is incompatible with the chemical environment of the SDA. Candidate grouts for the SDA, as discussed in Section 3.2, are composed of materials nonreactive with the SDA soil and have relatively low solubility in SDA soil water under the ambient pH and Eh conditions.

4.3.1 Cracking

Currently, the effect of monolith cracking on contaminant release rate is unclear. Cracking is a potential concern with cementitious grouts; however, plastic, clay-based, or hydrocarbon grouts would reform quickly under the ambient pressure to close any cracks that developed during curing. Currently, data are not available to indicate what cracking would be expected in SDA grout waste forms. Frequency, aperture, and connectivity are all factors that would affect water movement through the monolith. Though water flow through a fractured matrix can be rapid, as observed in SDA basalt, the leach rate is not necessarily increased because the microencapsulated waste form still minimizes contact between infiltrating water and contaminants. Finally, because the SDA is in a region of precipitating calcite, some researchers expect that any cracks in a grout monolith would be filled quickly by mineral deposition (Weidner et al. 2000).

Grout shrinkage also can result in cracking, but the degree to which a particular grout might shrink is controlled by a number of variables, including set temperature and constraining conditions. The *Biodegradation of Grout, Contaminant Diffusion, Solubility, and Technical Review of the In Situ Grout Treatability Study* (Weidner et al. 2000) states: “Because of boundary conditions (cementitious materials crack more when constrained) and slow chemical reactions, none of the tests will (or can) indicate the full degree of cracking to be expected over long time periods in a full scale application.”

Cracking is a difficult parameter to measure, as techniques conventionally used to evaluate structural integrity (e.g., coring and cutting) have the potential to alter or create cracks in the matrix. If monolith fracturing is determined to be a significant concern at the SDA, a number of soft grouts (e.g., Waxfix or soft-TECT) are available that would be well suited for this application.

4.3.2 Dissolution

The most significant mechanism causing grout degradation is dissolution of grout materials by slowly infiltrating water. As noted before, hydrocarbon-based grouts are virtually unaffected by water, but cementitious grouts are more susceptible. Mineral dissolution is a complex, but relatively well-understood process. Migrating water travels slowly through the soil and grout matrix, dissolving and precipitating minerals. Thermodynamic data available for chemical species can be used to predict the stability of contaminants because of the concentration of other minerals in the water, and the Eh and pH of the water. Though further evaluations of specific grout formulations may be required during the remedial design phase, recent data on a range of grouts (Loomis et al. 2002) has been used to estimate a dissolution rate for cementitious grouts.

Estimating durability of a particular waste stabilization material is not a straightforward task. The site geology, hydraulic properties, groundwater composition, and other factors must be considered in addition to chemical properties of the waste and grout materials. The following discussion provides a conservative estimate of the durability of ISG waste forms based on the dissolution rate of the most abundant chemical elements in the grout, namely the chemical components aluminum, silicon, and calcium. This estimate assumes that other factors (e.g., changing and recrystallizing mineral structures within the grout material) are negligible compared to the rate of dissolution of the waste form.

The estimate is computed from leach rate data measured by the ANS/ANSI-16.1 leach procedure. This procedure is a standard test method designed to determine the release rate of contaminants from porous-media waste forms (e.g., cement-based grout) used to stabilize waste materials. The ANS/ANSI-16.1 procedure measures the dissolution rate of the contaminant of interest into a specified amount of demineralized water (i.e., pure water at standard temperature and pressure over specified time periods for a total of 90 days).

4.3.2.1 Computations and Results. Time required for complete dissolution of each of the grout materials was estimated for aluminum, silicon, and calcium, assuming a 1-m cube of waste form material and using data from the 43-day test interval. Given this volume and shape, the ANS/ANSI-16.1 leachability procedure requires that 600 L of demineralized water surround the grout cube at any given time and be exchanged at a flow rate of 14 L/day so that the entire 600 L is renewed every 43 days. Results of the computations indicate that thousands of years will be required for complete loss of material composing the grouted waste form. For example, the tank closure grout would require 313×10^3 years for complete aluminum loss, 34×10^3 for complete silicon loss, and 80×10^3 years for complete calcium loss. Salt stone grout data indicated 650×10^3 years for aluminum, 26×10^3 years for silicon, and 139×10^3 years for calcium. All tested grout materials had comparable material loss rates.

4.3.2.2 Discussion. The ANS/ANSI-16.1 procedure provides a conservative estimate compared to actual grout dissolution rates in the natural groundwater at the SDA. The SDA ground waters are saturated with respect to the mineral calcite (CaCO_3) and contain a high level of dissolved silica (suggesting that cristobalite is the mineral phase controlling silica solubility), whereas the ANS/ANSI-16.1 leach test specifies demineralized water, which remains unsaturated. In addition, rate of flow in the test procedure is equivalent to 14 L/day for examples selected for computation and prevents the solutions from reaching saturation. The effect of composition difference between pore water and solvent water is illustrated by considering the Fick's law relationship given as $F = A(D_p \Delta C) / \Delta X$ (as described in Section

3) where F is the grout material flux, A is the area, D_p is the diffusion coefficient of material p , ΔX is thickness of the diffusion medium, and ΔC is the difference in concentration between pore water composition and the surrounding groundwater composition. In the SDA, ΔC is virtually zero and grout material flux would be virtually zero, indicating that material loss rates probably would be significantly slower than those used in the computations. Data indicate that all tested grout materials would provide mechanical stability and chemical buffering for thousands of years.

4.4 Grout Bench Scale Testing

Bench scale tests were designed to provide information about grout candidates for potential application of ISG at the SDA. The set of bench tests were applied to potential grout materials to determine their applicability to in situ long-term disposal, namely to measure (1) grout leach resistance to water, (2) effect of waste matrix on grout performance, (3) effect of grout on VOC retention within treated waste, and (4) grout chemical properties. The University of Ohio, Akron, conducted testing on a suite of grout samples under the direction of the INEEL (Loomis et al. 2002). Data results are used here to support the evaluation.

4.4.1 Diffusion Coefficient and Leachability Index Results

The bench leach tests were conducted to obtain data to evaluate the release rate of waste components from grout materials and to evaluate the effect of waste materials (e.g., organic sludge, nitrate salts, and INEEL soil on grout performance).

The leachability index is a numerical score used to compare retention of nonvolatile waste components within porous waste-form materials (e.g., grout) when leached by demineralized water. The leachability index is the negative exponent of the effective diffusion coefficient of the chemical specie of interest. The effective diffusion coefficient of a selected chemical specie is measured using a standardized batch leaching methodology (i.e., ANS/ANSI-16.1).

The effective diffusion coefficient and leachability index of calcium, aluminum, silicon, strontium, and nitrate was measured for each grout material. Results indicated that all of the tested grout formulations are leach resistant. For example, the nitrate leachability index ranged from 8.8 (American Minerals grout) to 11 (TECT grout), indicating that this highly soluble and mobile component is contained effectively within all the tested grout materials. (A typical leachability index for nitrate in water is approximately 5 [Weidner et al. 2000].)

The bench test showed that organic sludge (9 wt% loading), nitrate salt (12 wt% loading), and INEEL soil (50 wt% loading) had no effect on grout leachability. The test measured the strontium effective diffusion coefficient and leachability index for each mixture of interfering material and each grout. In all cases, leachability results virtually were unchanged from the values for pure grout.

4.4.2 Hydrogen Potential and Oxidation-Reduction Potential

Chemical properties of the grout material may affect and be affected by chemical properties of the waste site and waste materials. Acid-base properties (pH) and Eh of the leachates were measured during leach tests described above. All grouts produced alkaline, moderately oxidizing solutions having a pH in the range 10.9 (tank closure grout) to 11.4 (TECT), and an Eh of about 225 millivolts (mv) (S) to 390 mv (U.S. Grout). For comparison, groundwater at the SDA is slightly alkaline (i.e., approximately 7.16 pH), moderately oxidizing, and is in equilibrium with calcite and variable CO_2 soil gas concentration (Weidner et al. 2000).

5. IMPLEMENTABILITY

Implementing ISG technology is relatively well understood. A variety of grout products are available commercially and some bench and pilot scale testing have been completed. Past performance of ISG is well documented because a number of different grouting techniques have been tested and employed successfully across the DOE complex. Issues of worker safety are complicated by the unique hazards of the SDA, but subsurface application minimizes potential for worker exposure.

5.1 Grouting Techniques

The ISG term is used to describe a variety of techniques that apply stabilizing agents to the waste site. The techniques differ by the means of mixing, grout type, and pressure under which grout is applied.

5.1.1 Mechanical Mixing

Grout can be applied to contaminated soil sites by mixing grout into the surface soil with large augers or excavators. This technique, as implemented at the DOE Savannah River Site, is discussed in more detail in Appendix B, "In Situ Grouting Case Studies." This technique is reliable and ensures thorough mixing of the grout and contaminated soil. However, deeply buried waste at the SDA waste could not be mixed with conventional methods, and stirring the waste from the surface would pose unacceptable hazards. The presence of drums and other debris waste would preclude the use of mechanical mixing.

5.1.2 Permeation Grouting

Permeation grouting has been used widely in civil engineering and geotechnical fields. This technique uses boreholes drilled in the waste and cased with various types of perforated casing. In some applications, packers or sleeve pipes are used to inject grout at select depths. In these applications, low-viscosity grout is pumped into the borehole under low pressures (i.e., 20 to 70 psi [Dwyer 1994]).

Under low pressures, the grout fills all readily available voids in the waste and soil without significantly changing soil structure. Controlling the injection pressure prevents hydrofracturing of soil formations (Rumer and Ryan 1995). These low-pressure techniques are limited to formations with high permeability. Permeation grouting can be used with gravels and sands, but is not successful in silts and clays (Dwyer 1994).

This technique generally is not applicable to the SDA because of interstitial clay soil in waste pits. However, areas such as soil vaults may have void spaces around or beneath waste objects that could be filled with low-pressure grouting.

5.1.3 Injection Grouting

Injection grouting systems use high-pressure pumps and jetting nozzles to mix grouts with soil and waste. The high-velocity grout streams create a cutting and stirring action that brings the grout into intimate contact with the waste media, forming a relatively homogenous monolith. Rotation of the grouting lance or use of radial nozzles will create cylindrical columns that can be overlapped to effectively convert heterogeneous soil and waste into a monolithic soil-cement rock. Jet grouting can be used in a wide variety of soil types ranging from gravel to heavy clays (Mutch, Ash, and Caputi 1997). Multipoint injection and rotary-point injection (nondisplacement jet grouting) have been tested successfully for application to buried waste sites.

5.1.4 Multipoint Injection

Multipoint injection, a proprietary technique developed by Ground Environmental Services, uses a stationary lance with multiple injection nozzles to inject grout under high pressures. In previous demonstrations on simulated buried waste, cuttable polyvinyl chloride casing is driven into the formation with a direct-push rig similar to that used to install cone penetrometers. The lance is placed into the casing and grout is injected in lifts using a remotely located pumping system. The force of the grout cuts through the casing and into the waste matrix. This system also has been tested successfully for grouting sludge waste inside underground storage tanks at Oak Ridge National Laboratory (Kauschinger and Lewis 2000; Kauschinger et al. 2000).

The multipoint injection system relies on the interaction of multiple, high-speed monodirectional jets to mix the waste with various chemical agents. Instead of rod rotation, mixing occurs as multiple streams from the multipoint injection jets expand while they travel through waste. The use of high pressure (up to 11,000 psi) and an array of jets on the injection lance cause turbulent mixing, designed to uniformly mix waste with the grout. The multipoint injection techniques initially were devised to protect workers and pump equipment from becoming contaminated. With the multipoint injection process, this equipment is located in the uncontaminated support zone (ORNL 1996).

5.1.4.1 Rotary-Point Injection. Rotary-point injection, referred to as nondisplacement jet grouting, is a patented technology developed by the INEEL specifically to contend with SDA waste conditions. The nondisplacement jet grouting process involves mixing grout, soil, and waste debris at the subsurface level to form a large grout monolith (DOE 1999a; Loomis, Zdinak, and Bishop 1997). This technique accomplishes grouting without displacing contaminants or debris, or causing ground heaving. The overall site volume remains constant, while density of the site substantially increases.

In nondisplacement jet grouting, a modified well and coring drill rig is used to drive a 5-in.-diameter drill stem into the waste. The subassembly consists of two nozzles, 180 degrees opposed, approximately 12 in. from the cutting tip. A high-pressure positive displacement pump supplies grout to the drill stem and jet nozzles. Figure 2 provides an illustration of the system used previously at the INEEL.

The drill stem is driven into the waste using rotary-percussion action. A small flow of grout is pumped through nozzles for lubricating and temperature control as the stem is inserted. After the stem is driven to depth, the grout pressure is increased to approximately 6,000 psi. The drill stem systematically is withdrawn in discrete steps with two revolutions per step. The combination of withdrawal rate, rotation speed, withdrawal increment, and grout pressure is optimized for the waste area, ensuring that void spaces are filled completely, material or contaminant displacement is reduced, and grout returns remain minimal. The injection process is repeated on a tightly spaced triangular pitch grid (approximately 20 to 24-in. centers). Spacing of the injection points is designed to puncture every container (e.g., 55-gal drum). (Note: Waste containers in the SDA are up to 50 years old and expected to be largely deteriorated; therefore, close spacing would be recommended to ensure that contents of each drum are thoroughly mixed.)

This injection method has been shown to produce interlocking columns of grout under SDA conditions (Loomis, Zdinak, and Bishop 1997). Columns extend from the underburden soil up through the waste, terminating subsurface in the overburden. Interlocking columns cure into a solid monolith with no discernable edges between columns. Each waste container is filled from the inside as the drill stem and injection jets are raised back through the borehole. Cutting action of the jets fractures soil, plastics, wood, and other low-strength objects, and dislodges particles and small pieces of waste material, mixing them with grout and soil. Small amounts of liquids (i.e., oils and water) present in the waste are impacted by

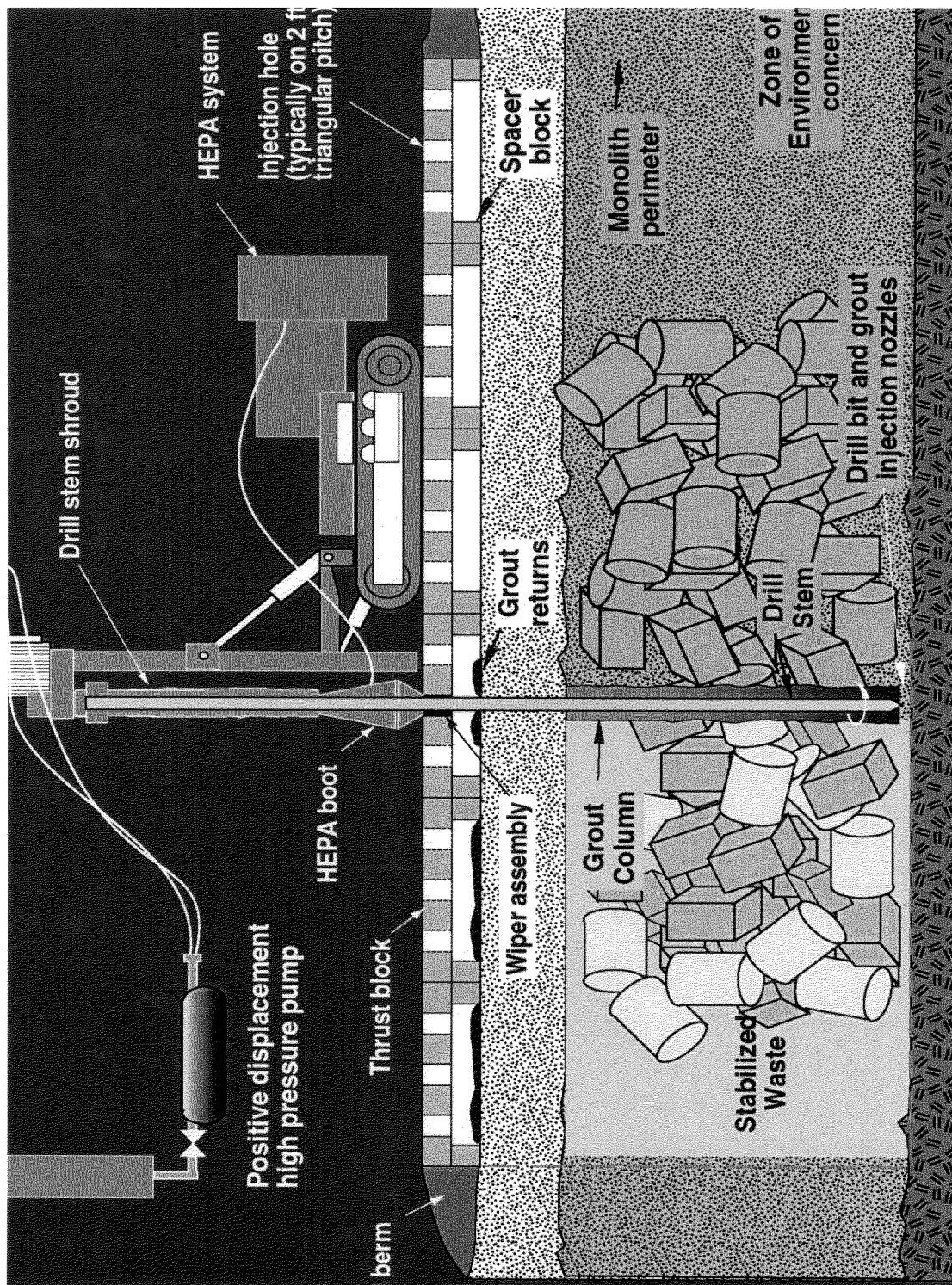


Figure 2. Schematic of the Idaho National Engineering and Environmental Laboratory injection grouting system.

the grout stream and dispersed over short distances in the grout. Large objects remain in place as the grout flows under pressure into voids around the objects. As a result, all readily accessible voids are filled (Loomis, Zdinak, and Bishop 1997).

5.1.5 Evaluation

Based on results of past field trials at the INEEL, injection grouting is the best-suited deployment system for ISG. Permeability grouting would be applicable only in areas with large void space or where waste was free of interstitial soil. Low porosity of soil and presence of containerized waste requires injection of grout at relatively high pressures and dense spacing.

Rotary point injection offers advantages of better mixing because jets are rotated and dwell time is controllable. Multipoint injection minimizes contamination control issues because injection lances are set in casings and can be left in place after grouting. The deployment system will need to be analyzed during the remedial design and optimized for specific waste streams. For the feasibility study evaluation, it is assumed that rotary-point injection would be used for pits and trenches where intimate mixing of waste and grout is desirable. In practice, design variations may be employed for specific areas (e.g., soil vaults) where disposable, leave-in-place injection lances and lower injection pressures may be better suited.

Past ISG work has deployed small drill rigs or direct-push units mounted on trucks or tracks. This equipment could be used in small or discrete areas within the SDA; however, in the large areas where thousands of injections would be required, it may be more efficient to use a more mobile system (Loomis et al. 2002, Appendix A). A gantry crane, fitted with the mast and hydraulic head from a small drill rig, may be used to inject grout. The crane would be operated remotely to position the injection lance over each hole. Pumps would be located remotely and no personnel would be required near the injection area during operations. To improve implementation, a tire-mounted crane would be used to eliminate the need for supporting rails. Using a wheel-mounted gantry crane for remediation was first proposed in 1996.^b Cranes are commercially available with 60-ft spans and suitable load capacities (Shuttlelift 2002). The injection mast would be moved across the span of the crane to inject grout in a linear array. After each row of holes is grouted, the entire crane is walked forward to the next row position. Such a deployment system would be expected to improve the safety and rate of operations.

5.2 Grout Availability

A wide range of grout types and formulations are available from commercial vendors. Many have been tested in bench-scale, field-scale, and actual remediation activities. For waste treatment applications, grouts have been developed to reduce site hydraulic conductivity, chemically bind certain contaminants in the grout crystal structure, and reduce the solubility potential of contaminants in infiltrating groundwater.

Site-specific performance goals should be used as the basis for selecting grout types because grouts need to be formulated for site-specific geochemical conditions and COCs. For example, at a site where diverting the flow of groundwater around a disposal trench is a goal, the hydraulic conductivity of a grout is an important parameter. At a more arid site, where the goal is to minimize the contaminant leach rate, a high-permeability, chemically reactive grout may be more effective.

b. Personal communication between A. T. Armstrong, DOE, and K. .M. Croft, INEEL, "Use of Gantry Cranes for SDA Remediation Work," 1996.

Grouts may be classified as particulate or cementitious grouts, chemical grouts, or a combination of both. Particulate grouts, the most common, typically are slurry mixtures of cement, bentonite, and water, often containing silicates and phosphates. Particulate cementitious grouts include Portland cements, Pozzolonic cements, high alumina cements, silica fume cements, and fly ash cements (Weidner et al. 2000). Cements commonly are modified with a wide range of additives. Chemical grouts typically contain a chemical base, a catalyst, and a solvent (e.g., water) (EPA 1998). Examples of chemical grouts include acrylate, urethane, and colloidal silica. Organic waxes, resins, and other hydrocarbon materials also may be classified as chemical grouts.

The variety of grout formulations available provides latitude to select the most appropriate formulations, considering remediation goals, waste types, and site-specific conditions. The following criteria (adapted from Grant et al. 2000) should be considered when identifying appropriate grouts for application at the SDA:

- Compatibility with injection-grouting techniques (including factors such as viscosity, density, solids suspension, cure and gel time, and past performance with jet grouting)
- Product safety (low cure temperature [i.e., less than 100°C], low toxicity, low neutron moderation)
- Environmental and geochemical compatibility with the SDA soil and groundwater (pH 8 to 13, Eh 600 to 200 mv)
- Available product performance data (contaminant-specific)
- Cost (material and production costs).

Engineering evaluations performed at the INEEL led to identifying a number of grout products that are strong candidates for use at the SDA (DOE 1999a), including:

- **TECT**—A pozzolonic cementitious grout with proprietary additives: TECT is a hematite analog grout with rheological properties that allow relatively small volumes of grout to disrupt and thoroughly mix with soil and debris, reducing the amount of grout required and spoils produced. In past studies, the grout successfully bound debris, oils, salts, mercury, and sludge into a low-permeability, ceramic-like product expected to have geologic durability similar to natural hematite (Carter 1998). TECT exhibited good performance in previous INEEL studies and was used in the remediation of the SDA Acid Pit (Loomis et al. 1998b). Tests conducted by Brookhaven National Laboratory showed the permeability of this grout (as a grout and soil mixture) to be less than 1×10^{-10} cm/second (Milian et al. 1997).
- **Tank closure grout**—Type V Portland cement, blast furnace slag, and silica fume grout developed at the Savannah River Site: This mixture specifically was designed to immobilize uranium, plutonium, and other actinides in waste sludge remaining in storage tanks. Tank closure grout has been reformulated for better injectability and is marketed under the trade name GMENT-12.
- **Waxfix**—A proprietary paraffin-based grout previously tested at the INEEL (Loomis, Zdinak, and Bishop 1997): Waxfix is injected as a molten plastic. The patented grout has been shown to saturate simulated waste debris and solidify organic liquids. Ongoing studies are evaluating the use of a boron additive to mitigate concerns of neutron moderation by the high hydrogen content. Tests by Brookhaven National Laboratory showed the permeability of these grouts (as grout and soil mixtures) to be less than 1×10^{-10} cm/second (Milian et al. 1997).

- **U.S. Grout (ultra fine grout)**—A Pozzolonic cement with low viscosity and delayed set times that would be conducive to jet grouting.
- **American Minerals (phosphate)**—A phosphate grout currently under development in the private sector: The use of phosphate in grout has shown significant chemical fixation properties (Singh et al. 1997).

It is likely that multiple grout types and formulations will be applied to the various waste types found in the SDA. A series of bench tests using a variety of grouts and actual SDA waste types likely will be conducted to validate actual grout formulations during the remedial design phase.

5.3 Past Performance

Numerous case studies exist to evaluate field scale performance of ISG systems. Appendix B presents detailed summaries of work to date using ISG to stabilize or contain radioactive waste sites. Studies include the following:

- **Oak Ridge National Laboratory WAG 4, Seeps 4 and 6 Project:** A multiphase, multistage, low-pressure permeation grouting in unlined radioactive waste disposal pits.
- **INEEL 1987 Grouting (Simulation) Study:** Evaluation of an experimental grouting process developed by Rockwell Hanford Operations that employed an I-beam to inject grout while simultaneously compacting the waste.
- **INEEL Acid Pit Project:** A treatability study of subsurface stabilization conducted in two phases (cold and hot testing) in a pit with radiological contamination and hazardous constituents.
- **Savannah River Site Old F-Area Seepage Basin:** A soil solidification remedy selected for an unlined seepage basin containing radiological and nonradiological contamination, with uranium as the primary risk driver (ORNL 1997).
- **Hanford Site Close-Coupled Barrier Demonstration:** A full-scale test of close-coupled barrier technology conducted on a buried 5,000-gal steel tank to evaluate integrity of the barrier created by the technique.
- **Brookhaven National Laboratory Glass Pit Remediation Technology Demonstration:** Application of the close-coupled technology to an actual remediation site involving unlined pits used for disposing of contaminated glassware and laboratory chemicals.
- **INEEL FY 1994 Innovative Grout Retrieval Demonstration:** A combination jet grouting and retrieval demonstration performed on a simulated waste pit constructed with 55-gal steel and cardboard drums and cardboard boxes filled with a wide range of simulated waste.
- **INEEL FY 1996 Innovative Grout Subsurface Stabilization Project:** A series of applied research tests of grouting techniques on simulated buried waste pits to evaluate the practicality of jet grouting as well as to establish hydraulic conductivity data for grouted and ungrouted sites.

No previous remedial actions match the SDA in size and variety of waste; however, the information provided by these case studies demonstrates that ISG technology is an effective solution with applications for the SDA. Technological methods and options that demonstrated success offered a basis for the evaluation of similar implementation at the SDA, while challenges confronted and lessons learned throughout the demonstrations and studies helped identify issues that may require consideration should the technology be selected for the SDA.

6. WORKER SAFETY

Any intrusive action taken at the SDA potentially poses a risk to remediation workers. Specific issues, discussed below, will need to be evaluated and mitigated during the remedial design phase, if ISG is a selected alternative.

6.1 Industrial Hazards

Heavy equipment used during grouting operations (e.g., drill rigs, cranes, high-pressure pumps, and batch plant equipment) pose significant industrial hazards. During remedial design and readiness activities, remediation workers will ensure that all systems are properly designed and meet appropriate engineering specifications and standards. Operations will be conducted in a planned and controlled manner with adequate procedures and trained crews to ensure the safety of workers involved. In addition, grout products that are flammable, toxic, or have cure temperatures above 100°C should be avoided. Suitable materials that do not have these characteristics have been identified by the INEEL (DOE 1999a).

6.2 Underground Fire or Explosion

The SDA waste includes nitrate salts and organic material (e.g., oils, graphite, and paper). The potential that these incompatible types of waste could become mixed and react to cause an underground fire or explosion was investigated previously (Beitel et al 1999; Quigley 1999; Dick 2001). In 1999, DOE commissioned an independent technical review panel to evaluate driving steel casings into Pit 9 using a rotary sonic drill rig. Through analysis and testing, the panel concluded that the risk of explosions or fires from sonic drilling was beyond extremely unlikely if the moisture content is greater than 5 wt% and the soil drill interface temperature is maintained below 150°C (Thompson et al. 2000). Though a thorough hazard evaluation will have to be conducted during the remedial design, for purposes of the feasibility study it is assumed that applying grout with its high moisture content will mitigate any potential for nitrate and organic reaction.

6.3 Criticality

Waste in the SDA includes process by-products from nuclear weapons component manufacturing. Though great care was taken to avoid accumulations of fissile material, evidence shows that some waste drums were overloaded (i.e., contain greater than 380 g of Pu-239 fissile gram equivalent). East (1995) identified 13 overloaded drums in stored waste at the RWMC. This information led to the assumption that a fraction of buried drums are similarly overloaded. A preliminary criticality safety evaluation assessed both cementitious and hydrocarbon-based grouts. Results indicate that no criticality hazards exist for cementitious grouts. However, the neutron moderating capabilities of paraffin grout posed a potential issue. Therefore, an administrative control was identified to ensure criticality safety when grouting with hydrocarbon-based grouts (i.e., B-10 would be added at 1.00 g/L to paraffin grout [Slate 2000]). However, during recent bench scale tests, it was difficult to maintain uniform distribution of boron in paraffin grout because the boron tended to settle to the bottom of the sample (see Section 4.4). If paraffin grout were selected during the remedial design, additional work would be required to ensure uniform distribution of boron. Alternatively, its application may be limited to areas with low fissile mass (e.g., organic Series 743 sludge).

6.4 Hazard Categorization

A preliminary hazard analysis has been performed for ISG application in the SDA (Peatross 2001). Though a final hazard analysis would need to be performed as part of the remedial design, the preliminary hazard analysis provides a basis for the feasibility study evaluation.

The preliminary hazard analysis used a systematic process to identify and assess hazards associated with hypothetical operation of ISG. Potential operational, external, and natural phenomena events that can cause accidents during operations were assessed. Though the preliminary hazard analysis document describes a conceptual design for an elaborate thrust block and drill string enclosure, the hazard evaluations were based on unmitigated scenarios as required by Standard (STD) DOE-STD-3009-94, "Preparation Guide for U.S. DOE Nonreactor Nuclear Facility Safety Analysis Reports."

Radiological and hazardous material inventories of the SDA were analyzed and bounding source quantities established. For example, it was assumed that bounding cases of drums containing 1,500 g of weapons-grade plutonium and 70 g of Am-241 would be encountered during ISG operations. From the total inventories, quantities of material at risk were developed for ISG operations.

Preliminary hazard analyses indicate that the potential amount of contamination brought to the surface would be minimal. Furthermore, because of encapsulating properties of the grout, contamination would not become easily airborne. Based on results of the analysis, unmitigated hazards are not expected to exceed dose evaluation guidelines established in U.S. Department of Energy Idaho Operations Office (DOE-ID) Order 420.D, "Requirements and Guidance for Safety Analysis." The ISG operation would not be classified as a nuclear operation in accordance with DOE-STD-1027-92, "Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports." Using the process established in DOE Standard DOE-EM-STD-5502-94, "DOE Limited Standard Hazard Baseline Documentation," and DOE-ID Order 420.D, the ISG operation is expected to be classified as a low radiological hazard.

Because of these analyses, ISG is not expected to be subject to many of the controls and processes associated with nuclear operations as some other remedial alternatives would be. Worker safety aspects of ISG would be governed under an extensive health and safety plan prepared in accordance with 29 CFR 1910.120 and 1926.65, "Hazardous Waste Operations and Emergency Response." The health and safety plan would include a detailed hazards analysis and identify engineering and administrative controls to ensure protection of workers.

6.5 Radiation Engineering Controls

Though worker risks in terms of evaluation guidelines are relatively low, the practical issue remains of controlling the spread of radioactive contaminants during and after grouting. Radiological control requirements (derived from 10 CFR 835, "Occupational Radiation Protection") will have a significant impact on the design and operation of ISG (as with any intrusive alternative at the SDA). Radiation safety experts at the INEEL agree that some form of surface control will be required to prevent the spread of contamination. However, techniques and equipment needed to control contamination will have to be evaluated and approved.

Previous INEEL tests on simulated waste have used concrete or steel platforms (i.e., thrust blocks) to cover the ground and contain grout returns. A flexible plastic bag or shroud encasing the drill string itself (i.e., drill-string enclosure) also has been tested for minimizing the potential for contamination spread. Though the system of thrust blocks and a drill string enclosure may be a viable approach, a

number of operational problems have precluded a successful demonstration. The adequacy of contamination controls will have to be evaluated and approved.

For the feasibility study, it is recommended that ISG be evaluated as if it were to be conducted inside a negative-pressure radiological confinement building. Functional requirements and specifications for the building would need to be developed during the remedial design, but it is assumed that the confinement would be a modular steel building erected in linear sections to allow the ISG system to progress down long rows. The structure would be maintained under negative pressure and ventilated through a high-efficiency particulate air filter system. The structure would be continually disassembled and moved as the ISG operation progresses across the SDA. While the pH indicated that potential for airborne contamination is very low, it is anticipated that the building would not become highly contaminated. A system of radiation monitors inside the structure would be used to verify that any contamination is maintained at acceptable levels. Workers would enter the building periodically to monitor contamination levels or to repair equipment, but would not be allowed inside during operations when the potential for surface contamination is highest.

After grouting operations, contaminated grout returns and spills potentially would remain exposed on the ground surface. Contamination would be considered fixed in grout; however, after the building is removed and the grout exposed to the environment, the material potentially could be weathered and blown by the wind to nearby facilities. To mitigate this, it has been suggested that post-operational grout returns be covered with a 3-ft layer of soil before the confinement building is moved. The soil would preclude erosion and possible airborne suspension of contaminants in the interim period before construction of the cap.

The approach to implementing ISG on a large scale with the SDA will need to be further evaluated and approved. Contamination control techniques have not been designed and demonstrated; therefore, uncertainties would need to be resolved by testing during the remedial design. In addition, because a final hazard analysis has not been performed, final worker risk calculations potentially would result in the ISG operation being classified as a nonreactor nuclear facility, requiring significant safety system structures and components to protect workers. In such an event, the design would need to be revised to meet specific quality and safety requirements.

7. PROBLEMATIC WASTE TYPES

7.1 Oil Waste Streams (Series 743 Sludge)

Series 743 organic sludge originating from the RFP (Section 2.2) contains high oil content, averaging 37 gal per 55-gal drum, and a grease-like consistency (Clements 1982). The Series 743 sludge was disposed of in large volumes distributed across multiple pits. Though it contains relatively small amounts of radioactive contaminants, the Series 743 sludge is a significant source of VOCs (e.g., carbon tetrachloride). Because researchers previously have had difficulty grouting oil-based waste (simulated Series 743 sludge [Loomis and Thompson 1995]), an evaluation of potential interference areas is required.

The ISG process fills the open space in buried waste with grout and intimately mixes the waste materials with grout. The result is a change in porosity and permeability of buried waste material and potential reduction of movement of volatile constituents out of the waste. To further assess the interference of organic oils on ISG, two sets of bench tests were performed at the University of Ohio in Akron, Ohio. Preliminary test results (Loomis et al. 2002) are summarized here.

In one scenario, grout is mixed (i.e., microencapsulated) with the waste matrix during the jet grouting process. This case was simulated by mixing 9-wt% organic oil with the grout and then measuring the release of VOCs into surrounding air after the mixture had cured.

In a second scenario, grout encloses a volume of waste material without mixing and acts as a mechanical barrier (i.e., macroencapsulation) to the release of the mobile organic contaminants. To simulate this case, an organic sludge was placed inside a cured cylinder of grout and the VOC migration was measured through approximately 1 in. of the cured grout barrier. Three grout compositions were tested (i.e., U.S. Grout, TECT, and tank closure grout). The organic sludge simulated the Series 743 sludge from the RFP and contained trichloroethylene, trichloroethane, tetrachloroethylene, and CCl_4 (Grant et al. 2000). Release of COCs was measured at 10-day intervals up to 90 days. As a standard for comparison, untreated simulated sludge material was allowed to off-gas and similarly was tested for VOC release.

Results showed that, in general, the three selected grout materials gave similar results in both the microencapsulation and macroencapsulation tests and that grout materials were effective in isolating organic contaminants. Between 0.02 and 0.08% of each VOC present was released into surrounding air in the test chamber during each 10-day test period. The tested grout reduced VOC release rates by four to five orders of magnitude compared to the untreated organic sludge. (Though grouting is expected to slow the release of VOC, final remedial decision may require pre-treatment of VOCs prior to grouting to be protective.)

Because of the macroencapsulating properties of ISG, pockets of oil (or other waste) are expected to be surrounded by competent grout. An individual drum of problematic waste may not be mixed thoroughly or properly cured, but it still would be effectively isolated from the environment. Therefore, areas with isolated drums of Series 743 waste would not be expected to reduce the effectiveness of ISG. Based on bench-scale test results, application of grout around individual Series 743 drums would, at a minimum, slow the release of VOCs. In addition, past work conducted at the Savannah River Site showed that specially formulated grouts incorporating blast furnace slag also could be used to destroy VOCs. The furnace slag creates a highly reducing environment in which the VOCs are degraded to nonhazardous components (ORNL 1996).

Though ISG is expected to be effective for low concentrations of organic waste, it is still unclear whether ISG would be effective in areas with high concentrations of Series 743 sludge. A review of the Series 743 sludge density distribution provided by Miller and Varvel (2001) indicates that the majority of the Series 743 sludge areas contain less than 2.5 drums/m² (see Figure 3). This would roughly equal 8% by volume oil (assuming a 14-ft deep waste zone and 37 gal oil/drum [see Section 2.2]). Based on past testing, it is reasonable to conclude that these areas would be effectively treated by ISG.

However, remaining areas with up to 12.5 drums/m² would be suspect because ISG has not been demonstrated with oil waste loading that high. Additional testing would be needed to demonstrate effective treatments, including reactive grout formulations, for these areas. Alternatively, a technology such as in situ thermal desorption could be applied to pretreat the high concentration oil areas before ISG.

In situ thermal desorption is a process for inserting an array of heated stainless steel pipe assemblies in the ground on an 8 × 8-ft spacing, to a depth of approximately 3 ft below the buried waste. Each assembly includes a sealed pipe containing an electrical-resistance heating element, a vented pipe used to extract gases, and thermocouples. Each extraction pipe would be connected to a pipe manifold conveying gases to an off-gas treatment system. The maximum temperature reached during in situ thermal desorption (i.e., 800°C), though well below the temperature at which soil and steel melt, is sufficient to destroy all VOCs.

The Series 743 sludge density map (see Figure 3) makes it evident that only a few areas (totaling less than 1 acre) would be considered interference areas. For the feasibility study, it is recommended that additional evaluation be done to ensure effective grouting or application of a pretreatment technology such as in situ thermal desorption.

7.2 Nitrate Waste Streams (Series 745 Sludge)

Series 745 sludge waste primarily is composed of dried sodium nitrate and potassium nitrate salts originating from evaporation ponds at RFP. High concentrations of salt compounds interfere with the curing of many cementitious grouts. However, recent bench-scale tests have demonstrated that waste loadings of up to 12-wt% nitrate have no effect on the grout leach resistance (see Section 4.4). Furthermore, past tests conducted at Oak Ridge National Laboratory demonstrated that high unconfined compressive strength and toxicity characteristic leaching procedure results could be achieved with waste loadings approaching 50 wt% nitrate salt^c (Spence et al. 1999).

As with the oil-based Series 743 sludge drums, intermittent Series 745 nitrate waste drums are not expected to significantly affect the performance of the ISG monolith. However, large caches of nitrate salt (e.g., in areas such as Pad A) may preclude effective curing of cementitious grouts. In these areas, chemically neutral polysiloxane or paraffin-based grouts have been shown to form an excellent microencapsulated waste form when mixed with granular nitrate waste (Loomis, Miller, and Prewett, 1997). A paraffin- or polyethylene-based grout likely also would be effective, but performance data specific to nitrate salts are not currently available. However, with these treatments salt waste is thoroughly mixed in an ex situ process to control the ratio of grout to waste and to ensure a consistent waste product. Using an in situ technique, as described in this report, has not been demonstrated on large caches of salt.

c. Researchers also have found a large number of grouts that failed when mixed with high concentrations of nitrate salts, and recommended that results not be extrapolated from one waste to another, but that testing on actual waste is needed before selecting a grout formulation.

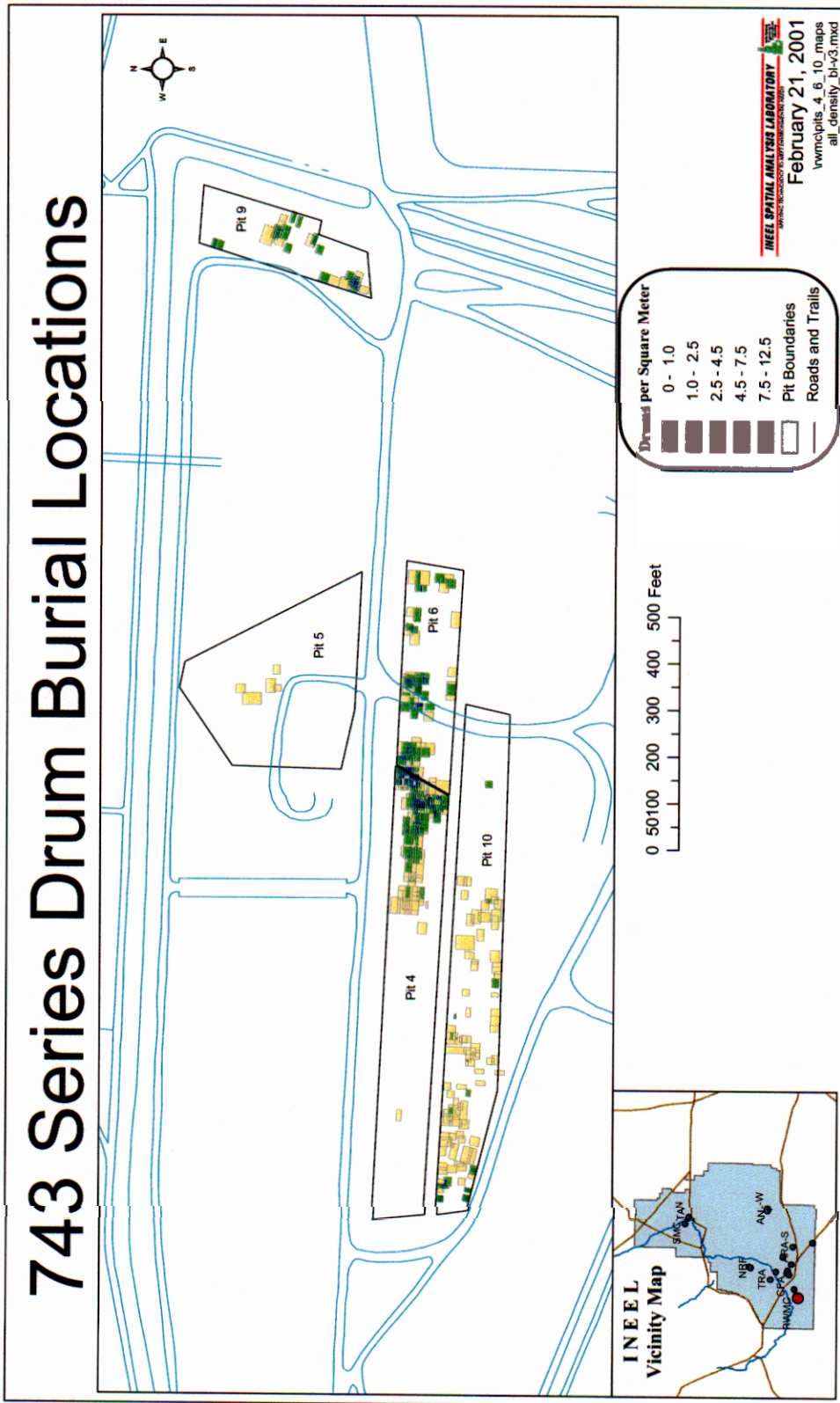


Figure 3. Density map of Series 743 sludge, adapted from Miller and Varvel (2001).

7.3 Large Objects (Concrete and Steel)

Injection-grouting techniques rely on advancing an injection lance (drill steel or well casing) through the waste with rotary-percussion action. Waste in the SDA includes construction debris (e.g. concrete, steel, and pipes) and large objects (e.g., trucks, tanks, and reactor vessel pieces). Intersecting such objects is likely to prevent full advancement of the injection lance and prevent grouting at that spot. If drill refusal is an isolated event, the offending object may be sufficiently encased with grout through adjacent holes. However, an area may become impossible to grout if a large cache of steel or other such debris is encountered.

Currently, maps are not available for areas containing large objects; therefore, it is difficult to predict whether drill refusal will be a significant problem. Many COCs are associated with waste (sludge) in drums that, as demonstrated by recent probing, are easily penetrated. From 1999 through 2002, more than 300 direct-push probes were inserted into a variety of waste streams in Pits 4, 9, and 10. All probes were inserted to underlying bedrock with no refusal from waste objects.^d Notably, the areas probed were selected because of their high concentrations of mixed TRU waste streams. Areas containing large caches of steel or demolition debris, vehicles, or other large objects may pose a challenge for thorough grouting of the waste layer.

d. A. T. Armstrong, North Wind Environmental personal communication with A. R. Baumer, INEEL, February 2001, "Probe Insertions."

8. RECOMMENDATIONS

Though ISG has been fairly well researched and has been implemented in limited cases, several uncertainties raise the risk of deployment at the SDA.

As described, reliable and consistent leach data do not exist for many SDA COCs. Use of leach data from available literature provides a sound basis for many assumptions used in the feasibility study (see Section 3); however, these data should not be relied on for design purposes because many COCs have not been addressed in previous work.

In addition, using the ANS/ANSI-16.1 leach test procedure is problematic, as discussed previously in this report. In reviewing this procedure, the current chair of the ANS/ANSI-16.1 working group commented that the test should be modified and rerun to produce data more representative of the SDA environment.^e Using deionized water, in equilibrium with air and with frequent changeout, is a worst-case scenario and is not representative of actual subsurface conditions. It is recommended that a static test be run to obtain leachability indexes and equilibrium distribution with the monolithic sample. Longer-term leach tests (multiple years) may also provide useful data. In addition, Eh and pH could be measured under an inert gas blanket to discern actual effect on water chemistry. Finally, simulated SDA water(s) could be tested to evaluate actual solubility potential in the SDA environment. It may also be beneficial to determine leach rates for actinides to develop a set of reliable data for SDA waste types.

During the development of the WAG 7 remedial investigation and feasibility study, soil vault rows and specific areas within the low-level waste trenches have been identified as potentially requiring treatment. The soil vault rows are unique in geometry and waste form; therefore, ISG implementation should be evaluated for soil vault rows with respect to the effective encapsulation of activated metal and other debris objects.

e. Spence, R. D., 2001, "Review and Critique of the OU 7-13/14 Draft Feasibility Study and Engineering Design Files," comments provided to BBWI Operation Review Board, October 31, 2001.

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